THE REUSE OF THE ALGERIAN PETROLEUM SLUDGE AS ALTERNATIVE RAW MATERIAL FOR CEMENT PRODUCTION

ALI BENLAMOUDI

A thesis submitted in
fulfillment of the requirement for the award of the
Doctor of Philosophy

Faculty of Civil and Environmental Engineering
University Tun Hussein Onn Malaysia

May 2019

For my beloved mother and father

ACKNOWLEDGEMENT

This research is a result of the grace surrounds me every moment, the day and the night, from the almighty Allah; I thank him for the power he gave me and for the guidance he blessed me. And I ask him that my work is purely for his sake and to be beneficial to me and to the whole Islamic nation.

I express my thanks to all my supervisors who helped me to achieve this research and gave me the right advices during both the months I spent in Algeria conducting my laboratory experiments and the period I spent at UTHM University in Malaysia. Without their guidance this PhD would not have been achievable.

I would like also to thank CRD-SONATRACH laboratories staff, who opened their doors to me to conduct the most important parts of my research. Special and warmest thanks go also to Dr. Khodja for his non-limited support to achieve this work starting from collecting the materials until conducting the laboratories test inside and outside CRD laboratories.

I am indebted to my beloved parents who supported me to continue my PhD studies, without whom I was so weak to accomplish this study. I would like to say a heartfelt and special thank to you my mom and my dad to everything you gave me to achieve this work, which is the fruit of your kind and non-limited support. May Allah bless you my mom and my dad the best rewards you deserve.

I would especially like to express my gratitude and warmest thank to my beloved wife who was accompanying me during my study abroad and was giving me the suitable support at every moment I was needed. Without you my soul, I would never finish this work and I would never find the courage to overcome all the issues during my PhD journey. Alhamdulillah, now everything is done and the certificate is yours.

Finally, but not least, special thanks to my family members and friends who supported me and helped me during my PhD studies, May Allah bless you all.

ABSTRACT

Recently, researchers are trying to incorporate alternative fuels and raw materials (AFR) to produce different types of cement like Ordinary Portland Cement (OPC), Portland Pozzolana Cement (PPC), Low Heat Cement and others. This incorporation is to avoid harmful impacts of AFR and to produce alternative sustainable cement. In this study, petroleum sludge (PS) waste, which is a type of AFR, was assessed to be incorporated into cement production as a sustainable treatment method for this waste that could also provide a low cost production of cement. To achieve the assessment method, PS was characterized for its physical, chemical and gas emission features to provide the advantages of its use as an ingredient to decrease the consumption of raw materials as well as the fuel within the cement plants. After characterization, 1%, 2.5% and 5% of PS was blended with the cement raw materials. Then, the prepared samples were incinerated within an electric furnace in order to produce the cement. After that, the cement produced was subjected to different chemical, structural, morphological and other analyses. The findings through this research were promising and the incorporation of petroleum sludge into cement plant could provide a sustainable low cost treatment method for this waste. The results showed that incorporation of 2.5% of petroleum sludge into cement production was considered as optimum and could produce an adequate and an acceptable quality of cement. This incorporation was able to decrease 2.5% of the cement raw materials and provided 2.1% to 2.4% of the energy required to produce cement. This research provides a sustainable low cost treatment method of PS that offer ultimate elimination of this waste by offering a decrease in the raw materials and a decrease in the fuel needed to produce cement.

ABSTRAK

Dewasa ini, penyelidik mencuba untuk menggabungkan bahan api alternatif dan bahan mentah (AFR) untuk menghasilkan pelbagai jenis simen seperti Simen Portland biasa (OPC), Simen Portland Pozzolona (PPC), Simen Haba Rendah dan lain-lain. Penggabungan ini adalah untuk mengelakkan kesan-kesan merbahaya dari AFR dan untuk menghasilkan simen alternatif mampan. Dalam kajian ini, sisa enapcemar petroleum (PS), yang mana sejenis AFR, telah dinilai untuk digabungkan ke dalam pengeluaran simen sebagai kaedah rawatan mampan untuk sisa ini yang juga menyediakan pengeluaran simen berkos rendah. Untuk mencapai keadah penilaian, PS telah dicirikan fizikalnya, kimia dan ciri-ciri pelepasan gas untuk memberikan kelebihan penggunaannya sebagai bahan untuk mengurangkan penggunaan bahan mentah serta bahan api di dalam loji. Selepas pencirian, 1%, 2.5% and 5% PS telah dicampurkan bahan mentah simen. Kemudian, sampel yang disediakan telah dibakar di dalam relau elektrik untuk menghasilkan simen. Selepas itu, simen yang terhasil tertakluk kepada kimia yang berbeza, struktur, morfologi dan analisis-analisis lain. Penemuan melalui kajian ini adalah memberangsangkan dan penggabungan enapcemar petroleum ke dalam loji simen dapat menyediakan rawatan kos rendah yang mampan untuk sisa ini. Keputusan menunjukkan bahawa penggabungan 2.5% enapcemar petroleum ke dalam pengeluaran simen dianggap sebagai optimum dan boleh menghasilkan kualiti simen yang memadai dan boleh diterima. Penggabungan ini berupaya mengurangkan 2.5% bahan mentah simen dan menyediakan 2.1% kepada 2.4% tenaga yang diperlukan untuk menghasilkan simen. Penyelidikan ini menyediakan kaedah rawatan PS berkos rendah yang mampan yang menawarkan penghapusan muktamad sisa ini dengan menawarkan pengurangan dalam bahan mentah dan pengurangan bahan api yang diperlukan untuk menghasilkan simen.

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LIST OF SYMBOLS AND ABBREVIATIONS

%	-	Percentage
µL	-	Micro liter
t/ hour	-	Ton per hour
t/ day	-	Ton per day
AFR	-	Alternative Fuel and Raw Materials
Al_2O_3	-	Aluminum oxide
AN	-	Algerian Standard
APHA	-	American Public Health Association
AR	-	Alumina Ratio
ASTM	-	American Society of Testing and Materials
ATSDR	-	Agency for Toxic Substances and Disease Registry
BAT	-	Best Available Techniques
BGS	-	British Geological Survey
BOF	-	Oxygen Furnace Sludge
BREFs	-	Best Available Techniques (BAT) reference documents
BS	-	British Standard
C_1-C_n	-	n-alkanes
C_2S	-	Dicalcium silicate (belite)
C_3A	-	Tricalcium aluminate
C_3S	-	Tricalcium silicate (alite)
C_4AF	-	Tetracalcium aluminoferrite
Ca_2SO_4	-	Calcium sulfate
$CaCO_3$	-	Calcite
$CaCO_3$	-	Calcium carbonate
$CaMg(CO_3)_2$	-	Dolomite

CaO	-	Calcium oxide
CaO	-	Carbon oxide
f-CaO	-	Free lime content
CEC	-	Commission of the European Communities
CEM I	-	Ordinary Portland cement type I
CEM II	-	Ordinary Portland cement type II
CFR	-	Code of Federal Regulations
CH_3COOH	-	Acetic acid
CH_4	-	Methane
Cl_2H_2	-	Dichloromethane
CO	-	Carbon monoxide
CO_2	-	Carbone dioxide
CRT	-	Cathode ray tube waste
DDI water	-	Distilled De-Ionized water
DWTPS	-	Drinking Water treatment Plant Sludge
EIPPCB	-	the European Integrated Pollution Prevention and Control Bureau
ELM	-	Emulsion liquid membrane
FA	-	Ferroalumina
Fe_2O_3	-	Iron dioxide
FWCS	-	Flotation wastes of copper slag
FWR	-	Foundation for water research
GC	-	Gas chromatography
GCV	-	Gross calorific value
GHG	-	Greenhouse gases
GICA	-	Industrial Group of Algerian Cement
GJ/ton	-	Gigajoule per ton
H_2O	-	Water
HCN	-	Hydrogen cyanide
HNO_3	-	Nitric acid
HP-5ms	-	Column with ((5%-Phenyl)-methylpolysiloxane) from Agilent Hewlett-Packard Company

ICP-MS	-	Inductively Coupled Plasma Mass Spectroscopy
IEA	-	The International Energy Agency
ITDI	-	Industrial Technology Development Institute
IWSA	-	Industrial wastewater sludge ash
K^+	-	Potassium ion
K_2O	-	Potassium Oxide
K_2SO_4	-	Potassium sulfate
kcal/kg	-	Kilocalorie per kilogram
KCl	-	Potassium chloride
kg	-	Kilogram
KWh	-	Kilowatt-hour
LDS	-	Lime dried sludge
LOI	-	Loss on ignition
LSF	-	Lime Saturation Factor
$m_{1,2,3}$	-	Weights of samples
mb/d	-	Million barrels per day
MBM	-	Meat and bone meal
mg	-	Milligram
MgO	-	Magnesium oxide
min	-	Minute
MJ/kg	-	Megajoule per kilogram
ml	-	Milliliter
mm	-	Millimeter
MPa	-	Megapascal
MSWI	-	Municipal Solid Waste Incineration
NA	-	Algerian standard
N/A	-	Not Available
Na_2O	-	Sodium oxide
Na_2SO_4	-	Sodium sulfate
NaOH	-	Sodium hydroxide
NCV	-	Net calorific value of constant pressure

NH_3	-	Ammonia
NO	-	Nitric oxide
NO_x	-	Nitrogen oxides
O_2	-	Oxygen
OMF-46	-	Oil well code I Hassi-Messaoud, Algeria
OPC	-	Ordinary Portland cement
OPEC	-	Organization of Petroleum Exporting Countries
P^{5+}	-	Phosphorus ion
P_2O_5	-	Phosphorus pentoxide
PAH	-	Polycyclic aromatic hydrocarbons
PCBs	-	Polychlorinated Biphenyls
PCDD/Fs	-	Polychlorinated dibenzo-p-dioxins (PCDDs) and polychlorinated dibenzofurans
PFA	-	Pulverized fuel ash
pH	-	Hydrogen potential
PHCs	-	Petroleum Hydrocarbons
ppm	-	Parts per million
PS	-	Petroleum sludge
QEMSCAN	-	Quantitative Evaluation of Minerals by Scanning electron microscopy
RCA	-	Recycled Concrete Aggregates
RCRA	-	Resources Conservation and Recovery Act
RDF	-	Refuse derived fuel
RG	-	Red gypsum waste
RM	-	Red-Mud
RMA	-	Recycled Masonry Aggregates
s/c	-	sand/cement ratio
SCAEK	-	Cement Company of Ain EL- Kebira
Si^+	-	Silicon ion
SiO_2	-	Silica
SO_2	-	Sulphur dioxide

SO_3	-	Sulfur trioxide
SONATRACH	-	Hydrocarbon Research, Production, Transportation, Processing and Marketing Company- Algeria
SR	-	Silica Ratio
SRF	-	Solid recovered fuel
SVOCs	-	Semi-volatile organic carbons
TCLP	-	Toxicity Characteristics Leaching Procedure
TGA	-	Thermogravimetric Analysis
TiO_2	-	Titanium dioxide
TPH	-	Total petroleum hydrocarbon
UCS	-	Unified compressive strength
UNEP	-	United Nations Environment Program
USD	-	United States Dollar
USEPA	-	United States Environmental Protection Agency
USGS	-	The United States Geological Survey
UTHM	-	Tun Hussein Onn University Malaysia
VOCs	-	Volatile organic carbons
w/b	-	water/binder ratio
w/c	-	water/cement ratio
WBCSD	-	World Business Council for Sustainable Development
WBG	-	World Bank Group
WPS	-	Water purification Sludge
WPSA	-	Water purification sludge ash
XRD	-	X-ray Diffraction
XRF	-	X-Ray Fluorescence
θ	-	Theta (angles of incidence)

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CHAPTER 1

INTRODUCTION

1.1 Background

Petroleum sludge (PS) is referring to a thick, viscous (Prakash *et al.*, 2015) intractable mixture encountered during the crude petroleum exploration, production, transportation, storage and refining processes (Prakash *et al.*, 2015; OPEC, 2016). PS consists of mainly water, solids of the crossed geological formations during the drilling operation and hydrocarbons that are deduced from both the drilled petroleum itself and the substances used for the drilling operation. This waste is generated from the two main operations of petroleum production, namely drilling operations during exploration of crude and its treatment. PS comprises mainly of water, drilling fluid, substances used for drilling operations and solids of the crossed geological formation. The considerable amount of PS that is generated every year in the oil producer countries caused several disadvantages including the lost of large areas of nature due to their occupancy by the disposed sludge and posing serious threats to the environment and the human health. Concerns regarding the harmful pollution of PS have attracted tremendous research on it to save the environment by reducing its severity. For that, a variety of PS treatment methods have been developed, such as landfarming, incineration, solidification/stabilisation, solvent extraction, ultrasonic treatment, pyrolysis, photocatalysis, chemical treatment, and biodegradation (Hu, Li & Zeng 2013). Nevertheless, these methods are associated to several drawbacks such as the high treatment cost, the toxic gas emitted to the environment and the remaining ash associated to the treatment process. Since PS is considered as waste that could be used as raw materials and/ or fuel, it could be considered as type of alternative fuel and raw material (AFR) that could substitute partially or wholly the fossil fuel and the natural mineral resources, respectively, in a waste management scheme called co-processing (ITDA, 2008). AFR could be generated from industries as residuals over production operations or from refused items after being used. The quantity of AFR has increased due to increased industrialization to meet economic growth targets. Most of the AFR pose acute effects on health and environment through at least one of the four characteristics defined by the Code of Federal Regulations (CFR): ignitability, corrosivity, reactivity and toxicity (WBCSD, 2005). These AFR materials contain several types of poisonous heavy metals and hydrocarbons. Heavy metals may include zinc, lead, copper, nickel, chromium and mercury. All the heavy metals could pose a significant danger to organisms and the environment through inhalation and leakage into the groundwater if the AFR materials are not treated before being disposed of in landfills. Hydrocarbons like gasoline, lubricating oil, motor oil, mineral spirits, lighter fluid/naphtha, lamp oil, and kerosene (Mowry *et al*., 2014) also have severe negative impacts that should be mitigated or contained.

On the other hand, in building and construction, cement is considered as one of the most important materials, which is used to maintain all the components together. The production of cement is raised enormously during the last decades because of the urban development (Chen, Hong & Xu, 2015). However, cement production is a very energy-intensive industry. Moreover, industries are facing major challenges regarding the number of energy resources invested in the production process, as well as the greenhouse gases emitted and air pollutant (Uwasu, Hara & Yabar, 2014). Therefore, due to the potential of PS, it could be investigated to be used as an alternative fuel to supply the cement kiln. Also, inorganic ashes remaining from the burning of sludge could be incorporated into molten clinker phases to reduce the materials required for cement manufacture (sand, clay, iron ore and limestone) into some extent. More advantage of this application over conventional combustion may include the destruction of any dioxins or furans produced during combustion due to the high kiln temperatures involved (Rodríguez *et al*., 2013). In addition, the potential reduction in CO_2 emissions from cement manufacturing is an advantage due to lower carbon content in PS relatively to fossil fuel (UNEP, 2010).

This study is an attempt to incorporate PS waste into cement production. This waste was investigated as an alternative fuel that could be used to supply the cement kiln with the emitted gas from burning of this waste within the cement kiln. Thus, the fuel needed to produce cement could be reduced. Furthermore, the ash that would be produced as a residue through the burning process of PS could be incorporated as an additive material with the cement raw materials. Thus, incorporation of PS could also reduce the raw materials amount needed for the cement production.

1.2 Problem statement

Oil companies generate toxic pollutants towards the environment in terms of air, water and soil. This is because a huge quantity of petroleum sludge (PS) waste is produced from the petroleum industry during its crude oil exploration, production, transportation, storage, and refining processes (Hu *et al.*, 2013). PS is considered as recalcitrant hazardous waste that may adversely affect the environment and the living things in case of improper disposal. Nevertheless, since PS contains high amount of hydrocarbons and heavy metals, recycling of this waste can be used as source of energy and source of raw material that provide an economic added value for different industries (Huang *et al.*, 2017). In Algeria, one typical drilling well generates around 1500 m^3 of PS. By counting more than 200 wells perforated every year, the generated PS would result in a quantity of around 300000 m^3, which is a huge amount. Therefore, in this context, this waste needs to be eliminated from the environment to avoid the recalcitrant sources of pollution that are accumulated especially hydrocarbons and heavy metals as they present the major sources of PS danger. Disposing of PS without treatment leads to migration of its toxic components into surface runoff and groundwater; thus, affecting the living organisms. In addition, the increasing amount of PS occupies hundreds of hectares that lead to significant danger zones (Ling & Isa, 2006). Hence, an elimination process or a treatment plant of PS is very important to eliminate this waste.

On the other hand, the demand for cement shows an increase during the last few years due to the importance of this material in building and construction. In Algeria, the first company of cement, which named Industrial Group for Algerian Cement (GICA), produces more than 10 million tonnes of cement every year (GICA, 2017). This production is associated with several issues such as the natural resources depletion due to the high demand as well as the usage of vast amounts of fuel. These disadvantages justified the need of sustainability in cement production (GICA, 2017).

However, previous studies have used different methods to treat PS, such as incineration, stabilisation/solidification, oxidation, biodegradation and others. However, all these methods are associated with various disadvantages and resulted in limited efficiencies towards some sources of danger (Hu *et al.*, 2013). It was reported that burning of PS is a promising method, which offers complete combustion of this waste and provides an alternative source of fuel that could be used for different purposes. Nevertheless, the generated ash through burning of PS and the expensive cost are the major shortcomings of this treatment method; thus, burning of PS needs more improvements. One of the alternative solution is to use this waste as an ingredient with the cement raw materials, which is leading to the elimination of hydrocarbon content and the encapsulation of the remained ash within the cement matrices formed (UNEP, 2010). In addition, PS will not require extra fuel or energy to be treated.

Due to very limited studies on the reuse of PS to produce sustainable cement, the focus of this research is to determine the efficiency of incorporating PS into cement production by taking the advantages to fully utilise this waste as a supplementary source of energy that will reduce the fuel consumption within the cement kiln and to reduce the raw materials needed for cement production. In addition, this study aims to eliminate the hydrocarbons and the heavy metals that are associated to PS.

1.3 Aim and Objectives

The aim of this research is to eliminate PS from the environment by incorporating it into cement production. For that, the objectives of this research are:

1. To characterize petroleum sludge (PS) in terms of heavy metals and hydrocarbons;
2. To assess PS as fuel support to supply the cement plant by identifying the parameters affecting its incorporation;
3. To identify PS as alternative raw material by its incorporation into cement clinker formation;

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